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TRANSMITTAL FORM (to be used for all correspondence after initial filing)	Application Number	10/702,440	
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	First Named Inventor	Fumito Nariyuki	
	Art Unit	1752	
	Examiner Name	Thori Chea	
Total Number of Pages in This Submission	15	Attorney Docket Number	FS-F03211-01

ENCLOSURES (Check all that apply)		
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Remarks Declaration should be considered along with Applicant's RCE submission dated November 28, 2005.		

SIGNATURE OF APPLICANT, ATTORNEY, OR AGENT			
Firm Name	TAIYO, NAKAJIMA & KATO		
Signature			
Printed name	Sheldon J. Moss		
Date	December 1, 2005	Reg. No.	52,053

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IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Appl. No.: 10/702,440
Applicant: Fumito Nariyuki
Filed: November 7, 2003
Title: PHOTOTHERMOGRAPHIC MATERIAL
Art Unit: 1752
Examiner: Thorl Chea
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Cust. No.: 37398

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SUBMISSION OF DECLARATION

Sir:

Applicant filed a "Submission For RCE" dated November 28, 2005 in response to the Office action of July 26, 2005. The submission was an Amendment. In the Remarks section of that Amendment, Applicant referred to a Declaration filed under 37 CFR 1.132, however, the declaration was not submitted therewith. Applicant now submits that declaration.

Respectfully submitted,

Sheldon J. Moss
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Date: December 1, 2005

THAT the experimentation set forth below was conducted by me or under my direct supervision.

EXPERIMENTS

Preparation of silver halide emulsions

< Preparation of silver halide (iodobromide) emulsion 1 >

A solution formed by adding 3.1 ml of 1 wt% potassium bromide solution to 1420 ml of distilled water and further adding 3.5 ml of sulfuric acid at 0.5 M/L concentration and 36.7 g of gelatin phthalide was kept at a liquid temperature of 30°C while stirring in a stainless steel reaction vessel, to which were added a solution A formed by diluting 22.22 g of silver nitrate with addition of distilled water to 95.4 ml and a solution B formed by diluting 15.3 g of potassium bromide and 0.8 g of potassium iodide with addition of distilled water to 97.4 ml volume, entirely, at a constant flow rate for 45 sec. Subsequently, 10 ml of an aqueous 3.5 wt% solution of hydrogen peroxide was added and 10.8 ml of an aqueous 10 wt% solution of benzimidazole was further added.

Further, a solution C formed by diluting 51.86 g of silver nitride with addition of distilled water to 317.5 ml and a solution D formed by diluting 44.2 g of potassium bromide and 2.2 g of potassium iodide with addition of distilled water to 400 ml volume were added such that the solution D was added entirely at a constant flow rate for 20 min, while the solution D was added by a controlled double jet (CDJ) method with pAg

being kept at 8.1. Potassium hexachloroiridate (III) was added by an entire amount so as to be 1×10^{-4} mol per 1 mol of silver 10 min after starting the addition of the solution C and the solution D. Further, an aqueous solution of potassium hexacyano ferrate (II) was added by an entire amount so as to be 3×10^{-4} mol per 1 mol of silver 5 sec after completing the addition of the solution C. pH was adjusted to 3.8 by using sulfuric acid at 0.5 mol/L concentration, stirring was stopped and a precipitation/desalting/water washing step was conducted.

pH was adjusted to 5.9 by using sodium hydroxide at 1 mol/L concentration, to prepare a silver halide dispersion at pAg of 8.0.

While keeping the silver halide dispersion at 38°C under stirring, 0.34 wt% of 1,2-benzisothiazolin-3-on in a 5 ml methanol solution was added and the temperature was elevated to 47°C. 20 min after the temperature elevation, sodium benzene thiosulfonate in a methanol solution was added by 7.6×10^{-5} mol based on 1 mol of silver and, further 5 min after, tellurium sensitizer C in a methanol solution was added by 2.9×10^{-4} mol per 1 mol of silver and aged for 91 min.

0.8 wt% of N,N'-dihydroxy-N'' diethyl melamine in a 1.3 ml methanol solution was added and, further 4 min after, 5-methyl-2-mercapto benzoimidazole in a methanol solution was added by 4.8×10^{-3} mol per 1 mol of silver, and 1-phenyl-2-heptyl-5-mercapto-1,3,4-triazole in a methanol solution was added by 5.4×10^{-3} mol per one mol of silver, to prepare a silver halide emulsion 1.

Particles in the thus prepared silver halide emulsion were cubic silver iodobromide (AgI 3.5 mol. %) particles with an average sphere-

equivalent diameter of 0.040 μm and a fluctuation coefficient for the sphere-equivalent diameter of 18%. The particle size, etc. were determined based on the average for the particles by the number of 1000 using an electron microscope. The resultant cubic silver iodobromide had an average γ -phase ratio of 0 %.

< Preparation of emulsion for coating solution A >

The silver halide emulsion 1 was dissolved, and benzothiazolium iodide in an aqueous 1 wt% solution was added by 7×10^{-3} mol pre 1 mol of silver. Further, water was added such that the content of the silver halide as silver was 38.2 g per 1 kg of the emulsion for coating solution.

< Preparation of silver halide (iodide) emulsion 2 >

A solution formed by adding 4.3 ml of a 1 wt% potassium bromide solution to 1420 ml of distilled water and further adding 3.5 ml of sulfuric acid at 0.5 mol/L concentration and 36.7 g of gelatin phthalide was kept at a liquid temperature of 42°C while stirring in a stainless steel reaction vessel, to which were added a solution A formed by diluting 22.22 g of silver nitrate with addition of distilled water to 195.6 ml and a solution B formed by diluting 21.8 g of potassium iodide with addition of distilled water to 218 ml, entirely, at a constant flow rate for 9 min. Subsequently, 10 ml of an aqueous 3.5 wt% solution of hydrogen peroxide was added and an aqueous 10 wt% solution of benzimidazole was added by 10.8 ml.

Further, a solution C formed by diluting 51.86 g of silver nitrate with

addition of distilled water to 317.5 ml and a solution D formed by diluting 60 g of potassium iodide with addition of distilled water to 600 ml volume were added such that the solution C was added entirely at a constant flow rate for 120 min while the solution D was added by a controlled double jet method with pAg being kept at 8.1. Potassium hexachloroiridate (III) was added by an entire amount so as to be 1×10^{-4} mol per 1 mol silver 10 min after starting the addition of the solution C and the solution D. Further, an aqueous solution of potassium hexacyano ferrate (II) was added by an entire amount so as to be 3×10^{-4} mol per 1 mol of silver 5 sec after completing the addition of the solution C. pH was adjusted to 3.8 by using sulfuric acid at 0.5 mol/L concentration, stirring was stopped and precipitation/desalting/water washing step was conducted. pH was adjusted to 5.9 by using sodium hydroxide at 1 mol/L concentration, to prepare a silver halide dispersion at pAg of 8.0.

While keeping the silver halide dispersion at 38°C under stirring, 0.34 wt% 1,2-benzisothiazolin-3-on in a 5 ml methanol solution was added and the temperature was elevated to 47°C. 20 min after the temperature elevation, sodium benzene thiosulfonate in a methanol solution was added by 7.6×10^{-5} mol based on 1 mol of silver and, further 5 min after, a tellurium sensitizer B in a methanol solution was added by 2.9×10^{-4} mol per 1 mol of silver and aged for 91 min.

0.8 wt% of N,N'-dihydroxy-N'' diethyl melamine in a 1.3 ml methanol solution was added and, further 4 min after, 5-methyl-2-mercapto benzoimidazole in a methanol solution was added by 4.8×10^{-3} mol per 1 mol of silver, and 1-phenyl-2-heptyl-5-mercapto-1,3,4-triazole in a

methanol solution was added by 5.4×10^{-3} mol per one mol of silver, to prepare a silver halide emulsion 2.

Particles in the thus prepared silver halide emulsion were tetradodecahedral pure silver iodide particles with an average sphere-equivalent diameter of 0.040 μm and a fluctuation coefficient for the sphere-equivalent diameter of 18%. The particle size, etc. were determined based on the average for the particles by the number of 1000 using an electron microscope. The resulted silver iodide had an average γ -phase ratio of 30 %.

[Preparation of emulsion B for coating solution]

The silver halide emulsion 2 and benzothiazolium iodide as an aqueous 1 wt% solution was added by 7×10^{-3} mol per 1 mol of silver. Further, water was added such that the content of the silver halide per 1 kg of the emulsion for coating solution was 38.2g.

1-(3-methyl ureido phenyl)-5-mercapto tetrazole was added so as to be 0.34 g per 1 kg of the mixed emulsion for each of the coating solutions A and B.

[Preparation of photothermographic materials 1 to 6]

Photothermographic materials 1 - 6 were prepared in the same manner as described in Example 1 of the specification except for using emulsions A or B for coating.

< Preparation of photothermographic materials 7 to 12 >

Photothermographic materials 7 to 12 were prepared in the same manner as for the Photothermographic materials 1 to 6 except for using the emulsion B for coating instead of the emulsion A for coating, with the coating amount of the silver halides being as described in Table 1.

Table 1

Photosensitive material No.	Silver halide composition *	Average sphere-equivalent diameter [μm]	AgX silver coating amount [g/m ²]	Sensitivity to developing time			Color difference to developing time (ΔE)		Color storability (ΔE)	Remark
				13sec	14sec	15sec	13sec	14sec		
1	AgBr96.5 I3.5	40nm	0.007	92	100	107	0.69	0.58	0.97	Comparative Example
2	AgBr96.5 I3.5	40nm	0.01	93	100	107	0.74	0.59	1.03	Comparative Example
3	AgBr96.5 I3.5	40nm	0.04	93	100	106	0.77	0.62	1.01	Comparative Example
4	AgBr96.5 I3.5	40nm	0.1	93	100	106	0.79	0.63	1.01	Comparative Example
5	AgBr96.5 I3.5	40nm	0.4	92	100	107	0.84	0.61	1.02	Comparative Example
6	AgBr96.5 I3.5	40nm	0.5	92	100	108	0.81	0.56	1.10	Comparative Example
7	AgI100	40nm	0.007	99	100	100	0.29	0.31	0.54	Invention
8	AgI100	40nm	0.01	98	100	101	0.34	0.38	0.64	Invention
9	AgI100	40nm	0.04	98	100	101	0.37	0.41	0.74	Invention
10	AgI100	40nm	0.1	97	100	102	0.41	0.44	0.87	Comparative Example
11	AgI100	40nm	0.4	95	100	103	0.47	0.51	1.02	Comparative Example
12	AgI100	40nm	0.5	92	100	107	0.84	0.74	1.13	Comparative Example

* Average γ-phase ratio of AgBr_{96.5}I_{3.5} particles was 0%.Average γ-phase ratio of AgI₁₀₀ particles was 30%.

As can clearly be seen from the results shown in Table 1, comparing with the photosensitive materials of Nos. 1 to 6 that use silver iodobromide (average γ -phase ratio: 0 %) and the photosensitive materials of Nos. 10 to 12 with high silver iodide content but having a silver content in the coating (more than 0.05 g/m^2), the photosensitive materials of Nos. 7 to 9 with high silver iodide content (average γ -phase ratio: 30 %) and having a silver content in the coating of 0.05 g/m^2 or less have unexpectedly reduced variation in sensitivity and color tone due to difference in developing time (13 seconds \rightarrow 15 seconds), as well as unexpectedly improved storability of color tone.

Further, comparing the photosensitive materials of Nos. 1 to 6, in which the amount of silver in the coating was reduced but no improvement in sensitivity was observed for the developing time of 13 seconds, to the photosensitive materials of Nos. 7 to 9 with a silver content in the coating of 0.05 g/m^2 or less, there is the unexpected result of enhanced sensitivity (ΔE) (improvement in progression of development).

Claims 1, 3, 7-10, and 12-20 were rejected under 35 U.S.C. 103 as being anticipated by U.S. Patent No. 6,143,488 to Uytterhoeven et al. in view of EP 01096310 (EP '310). Uytterhoeven does not explicitly describe the amount of silver in the coating either in the specification or the Examples. Therefore, the amount of silver has been calculated based on the formulation described in Examples.

US 6143488 (Uytterhoeven) Example 1:

First, the amount of AgI in the coating is determined:

10.7 % silver behenate (MW = 448) in 4g of silver behenate dispersion
 $= 4 \times 0.107 / 448 = \underline{9.6 \times 10^{-4} \text{ mole}}$... (1)

0.72 g of PC03 ... 8 mol. % with respect to silver behenate ... (2)

0.16 g of phthalazine

0.74 g of reducing agent (LOWINOXTM)

1.2 g of MEK

Total 6.82 g = 6.82 cc (when specific gravity is 1) ... (3)

Coating is carried out at a wet coating thickness of 80 μm ($80 \times 10^{-6} \text{ m}^3$ per 1 m^2)
= 80 cc/m² ... (4)

From (1), (3) and (4), the amount of silver in the coating is:

$$9.6 \times 10^{-4} \times 108 \text{ (MW of Ag)} \times 80/6.82 = 1.22 \text{ g/m}^2.$$

From (2), the iodine compound PC03 is 8 mol. % with respect to silver.

$$\text{The amount of silver of AgI in the coating} = 1.22 \text{ g/m}^2 \times 8 \% \approx \underline{0.1 \text{ g/m}^2}.$$

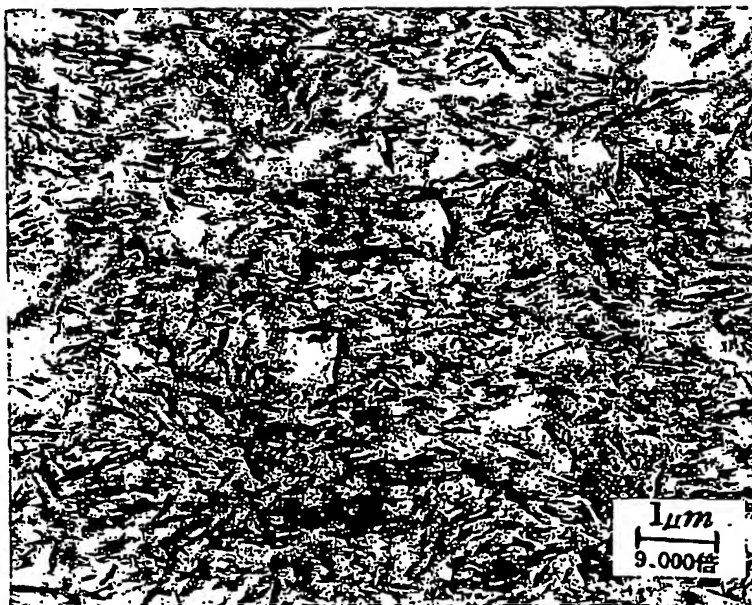
Therefore, the amount of silver of AgI in the coating calculated based on the formulation in the Examples of Uyterhoeven is substantially larger than the amount of silver, 0.005 to 0.05 g/m^2 , of the photosensitive silver halide in the coating required by claim 1 of the present application.

Further, the formulation in the examples of Uyterhoeven is is-situ silver halide prepared by conversion, in contrast to the requirements set forth in claim 1. There is a substantial difference between organic silver salt dispersions prepared through conversion and those in which a photosensitive silver halide is formed in a state where the non-photosensitive organic salt is not present.

Organic silver salts are prepared through stages of: organic acid \rightarrow alkali metal soap \rightarrow organic silver salt. Among them, only the alkali metal soap is hydrophilic and is stably dispersible in water. The organic acid and the organic silver salt are highly hydrophobic and cannot be stably dispersed in water, and therefore need to be dispersed

using an organic solvent. In contrast, silver halides that have been prepared in advance (preformed) are water dispersed using a hydrophilic colloid such as gelatin, and therefore are stably dispersible in water. The above explanation and Figs 1 and 2 (below) illustrate conditions of dispersion of a silver halide and an organic silver salt in an image forming layer.

Figure 1 shows a dispersion obtained by the process in which a preformed silver halide is present during formation of an organic silver salt (the present invention). In this process an alkali metal soap stably dispersible in water and a water-dispersed silver halide are mixed in water. Therefore, they are readily blended with each other and dispersed without forming aggregation. From this well-dispersed state, the alkali metal soap is converted into an organic silver salt by addition of a water soluble silver salt. Accordingly, this well-dispersed state can be maintained. In the mixture of the silver halide and the organic silver salt prepared by this type of process, silver halide particles do not aggregate with each other and do not directly contact the organic silver salt, and are observed as a uniform dispersion.



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Figure 1. Process in which a preformed silver halide is present during formation of an organic silver salt. Black regions are silver halide and light gray particles are the organic silver salt. It can be seen that the silver halide and the organic silver salt are uniformly dispersed.

Figure 2 shows a dispersion obtained by the conversion process (as described in the Examples of Uytterhoeven). The conversion process partially converts an organic silver salt into a silver halide by reacting the organic silver salt with a halogen ion. Accordingly, the formed silver halide is coupled to the surfaces of crystals of the organic silver salt, and therefore cannot be dispersed independently from the organic silver salt.

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Figure 2: Conversion process. Black portions are silver halide and light gray particles are the organic silver salt. Silver halide is distributed on crystals of the organic silver salt.

As can be seen from the above explanation and photomicrographs, the final products of these two types of dispersions are not the same. Further, the preformed silver halide according to the present invention can be used with conventional particle forming methods to adjust particle size, particle size distribution, halogen composition structure such as core shell and the like, and also can be used with techniques such as metal ion doping and chemical sensitization, whereas none of these techniques can be used with the conversion process (Figure 2).

I further declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

Tomoyuki Ohzeki
Tomoyuki OHZEKI

November 29, 2005
Date